GASEOUS SECONDARY ELECTRON DETECTION AND CASCADE AMPLIFICATION IN THE ENVIRONMENTAL SCANNING ELECTRON MICROSCOPE

By

Scott Warwick Morgan

A THESIS SUBMITTED IN FULFILMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY

FACULTY OF SCIENCE
UNIVERSITY OF TECHNOLOGY, SYDNEY
AUSTRALIA
2005

Certificate

I certify that the work in this thesis has not previously been submitted for a degree nor has it been submitted as part of requirements for a degree except as fully acknowledged within the text.

I also certify that the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

_____

Signature of Author

To My Family.

Acknowledgments

This work in this thesis was conducted under the supervision of Assoc. Prof. Matthew Phillips, director of the Microstructural Analysis Unit (MAU), University of Technology, Sydney (UTS). I would like to sincerely thank Assoc. Prof. Phillips for his undivided help and support during the entirety of my research. His cool, calm and collected, but rigourous, approach to science made my time spent with him very learned and enjoyable.

I would like to thank Dr Miloš Tóth, currently at the FEI company, Boston, for his ongoing help, fruitful discussions and experimental collaboration both at UTS and the Polymer and Colloids Group, Cavendish Laboratory, University of Cambridge. I would also like to thank the staff of the physics department at UTS for valuable discussions and for the loan of some of the equipment used in experiments. I wish to thank the staff at the MAU, consisting of Richard Wuhrer, Mark Berkahn and Katie McBean, for outstanding technical support and friendly advice.

I would especially like to thank my fiancé Larissa Lembke for her devoted love and support over the entire course of my PhD. I am grateful to Larissa for putting up with my 'occasional' bad moods, late nights, missing dinners and proof-reading this thesis. "Thanks darling".

I sincerely thank my parents (Warwick and Lynette), nanna (Pearl) for their utmost moral and financial support, and for continuously being there for me. I also gratefully thank the other close members of my family and friends for their help,

support, physics discussions and for sharing a beer with me when I needed it most. I apologize to anyone for whom felt at times my PhD was of more importance than them.

Lastly, I wish to thank Chris Cornell, James Hetfield and my Ibanez RG 450 for the jam sessions when my brain could not take anymore.

Table of Contents

List of Figures

List of Tables

Nomenclature

Table 1: List of symbols and abbreviations.

a_H	Bohr radius
A	atom
A_e	total electronic gas amplification
A_e^{BSE}	gaseous BSE electronic amplification
A_e^{PE}	gaseous PE electronic amplification
A_e^{SE}	gaseous SE electronic amplification
A_{hv}	photon amplification
Al_2O_3	alumina
Ar	argon gas
BSE	backscattered electron
BWD	bandwidth
c	vacuum speed of light

Table 1: Continued...

C	capacitance
C_{GSD}	GSD contrast
CL	cathodoluminescence
CO_2	carbon dioxide gas
d	sample-electrode separation
d_{eff}	effective gap distance
$\bar{d}_{BSE}$	average BSE path length
D	transition region distance (below PLA1)
DC	direct current
DP	diffusion pump
DR	dielectronic recombination
DSR	dissociative recombination
e	electron charge
e^-	electron
E	electric field strength
E_{ion}	ion electric field strength/space charge field strength
E_{net}	net electric field strength

Table 1: Continued...

E_{GSED}	GSED electric field strength
E/p	reduced electric field
EBIC	electron beam induced current
EC	environmental chamber
ESE	environmental secondary electron
ESEM	environmental scanning electron microscope
E-T	Everhart-Thornley
$f_e(\theta)$	scattering amplitude
FET	field effect transistor
g_m	metastable geometrical loss factor
g_p	photon geometrical loss factor
G_{PMT}	PMT gain
GSD	gaseous scintillation detector
GSED	gaseous secondary electron detector
GSI	greyscale intensity
h	Planks constant
h	transition region distance (above PLA1)

Table 1: Continued...

hv	photon
H	distance between PLA1 and PLA2
H_n	Struve's function of order n
H_2	hydrogen gas
H_2O	water vapour
He	helium gas
HFW	horizontal field width
I_e^{hv}	electron avalanche generated photon current
I_{ind}	induced current
I_{ion}	total ionization current
I_{BSE}^g	gaseous BSE ionization current (*ad infinitum* avalanches)
I_{BSE}^{hv}	total scintillation BSE photon current
$I_{BSE_0}^g$	gaseous BSE ionization current (single avalanche)
$I_{BSE_0}^{hv}$	primary scintillation BSE photon current
$I_{BSE_1}^{hv}$	secondary scintillation BSE photon current
I_{GSED}	GSED current
I_{PE}	PE beam current

Table 1: Continued...

I_{PE}^{g}	gaseous PE ionization current (*ad infinitum* avalanches)
I_{PE}^{hv}	total scintillation PE photon current
$I_{PE_0}^{g}$	gaseous PE ionization current (single avalanche)
$I_{PE_0}^{hv}$	primary scintillation PE photon current
$I_{PE_1}^{hv}$	secondary scintillation PE photon current
I_{PMT}^{in}	PMT input current
I_{PMT}^{out}	PMT output current
I_R	resistor current
I_{SE}^{g}	gaseous SE ionization current (*ad infinitum* avalanches)
I_{SE}^{hv}	total scintillation SE photon current
$I_{SE_0}^{g}$	gaseous SE ionization current (single avalanche)
IP	ion pump
IR	infrared spectrum
ISC	induced stage current
J	ionization energy
J_n	Bessel function of order n
k	Boltzmann constant

Table 1: Continued...

k	cascade amplification feedback factor
K	kinetic energy
K_i	ion kinetic energy
K_n	neutral atom/molecule kinetic energy
K_{SE}	SE kinetic energy
K_0	modified Bessel function of the second kind of zero order
L	dimensional unit of length
LED	light emitting diode
m	average number of scattering events
m_e	electron rest mass
M	metastable-cathodic electron emission probability
M	molecule
MGSI	mean greyscale intensity
n	number density/concentration
n_e	electron number density/concentration
n_i	ion number density/concentration
N_e^c	number of cathodic electrons

Table 1: Continued...

N_e^g	number of gaseous electrons
N_{hv}^g	number of gaseous photons
N_n	Neumann's Bessel function of the second kind of order n
N_p^c	number of cathodic photoelectrons
N_{BSE}^{hv}	number of photons generated by BSEs
N_{CL}^{hv}	number of photons generated by CL
N_{PE}^{hv}	number of photons generated by PEs
N_{SE}^{hv}	number of photons generated by SEs
N_T^{hv}	total number of photons
N_2	nitrogen gas
NO	nitrous oxide gas
O_2	oxygen gas
p	pressure
p_{max}	maximum efficiency pressure
p_{max}^e	maximum ionization efficiency pressure
p_{max}^{hv}	maximum excitation efficiency pressure
p_{Ar}	argon pressure

Table 1: Continued...

p_{H_2O}	water vapour pressure
p_{N_2}	nitrogen pressure
p_1	stagnation pressure
P	photoelectric yield
$P(x)$	collision probability
PCB	printed circuit board
PE	potential energy
PE	primary electron
PI	positive ion
PLA	pressure-limiting aperture
PMT	photomultiplier tube
Q	charge
r	radial distance
r_{ij}	molecular inter-atomic distance
r_0	minimum atomic distance
$r_{1/2}$	skirt half-width radius
R	correlation coefficient

Table 1: Continued...

R	effective atomic radius
R	resistance
R_m	maximum electron interaction range
R_L	load resistance
R_S	series resistance
RC	time constant
RC_{nf}	noise filter time constant
RDR	radiative dielectronic recombination
RE	resonant excitation
RP	rotary pump
RR	radiative recombination
s	particle displacement
$sk_e(\lambda)_p$	photocathode spectral sensitivity
S_{BSE}	BSE ionization efficiency
S_{PE}	PE ionization efficiency
SbKCs	Bailkaline
$(S/B)_{SE}$	SE signal-to-background ratio

Table 1: Continued...

$(S/B)^{e}_{SE}$	electronic SE signal-to-background ratio
$(S/B)^{hv}_{SE}$	electroluminescent SE signal-to-background ratio
SE	secondary electron
SEM	scanning electron microscope
SNR	signal-to-noise ratio
t	time
T	absolute temperature
T	dimensional unit of time
TBR	three-body recombination
TV	television
$T(\lambda)$	transmission response
UV	ultra violet spectrum
v	photon frequency
v_c	critical photon frequency
v_e	electron drift velocity
$\bar{v}_e$	average electron velocity
v_i	ion drift velocity

Table 1: Continued...

V	voltage
V_e^1	first excitation potential
V_i^1	first ionization potential
$V_p(\rho)$	plural scattering probability distribution
$V_s(r)$	single scattering probability distribution
V_{GSD}	GSD voltage
V_{GSED}	GSED voltage
V_{HT}	PMT high tension voltage
V_{PMT}^{out}	PMT output voltage
V_S	voltage signal
VIS	visible spectrum
VPSEM	variable pressure scanning electron microscope
VUV	vacuum ultra violet spectrum
WD	working distance
z	gap distance
z_Ω	maximum SE-ion recombination distance
Z	atomic number

Table 1: Continued...

α_{exc}	excitation coefficient
α_{ion}	first Townsend ionization coefficient SE/ESE ionization efficiency
α_{ion}^{sw}	first Townsend ionization coefficient SE/ESE ionization efficiency (swarm conditions)
γ	total second Townsend coefficient
γ_i	ion second Townsend coefficient
γ_m	metastable second Townsend coefficient
γ_n	neutral atom/molecule second Townsend coefficient
γ_p	photoelectron second Townsend coefficient
Γ_e	electron transit time
Γ_i	ion transit time
δ	SE emission coefficient
δ_{eff}	effective SE emission coefficient
Δ	reaction by-products
ε	electron energy
ε_{BSE}	BSE energy
ε_{PE}	PE energy
$\bar{\varepsilon}_{SE}$	average SE energy
ε_0	electron rest energy

Table 1: Continued...

ζ_T	mass thickness of transition region
$\zeta(t)$	SE-ion recombination rate
η	BSE emission coefficient
θ	scattering angle
λ	photon wavelength
λ_c	critical photon wavelength
λ_e	relativistic electron wavelength
λ_{SE}	SE inelastic mean free path
λ_{SE}^{e}	SE mean free path (ionization)
λ_{SE}^{hv}	SE mean free path (excitation)
λ_1	minimum photon wavelength
λ_2	maximum photon wavelength
μ_m	metastable absorption coefficient
μ_p	photon absorption coefficient
ρ	reduced radial distance
ρ	total SE-ion recombination coefficient
ρ_{DSR}	SE-ion recombination coefficient (DSR)
ρ_{RR}	SE-ion recombination coefficient (RR)

Table 1: Continued...

σ_e^s	elastic scattering cross section
σ_i^s	inelastic scattering cross section
σ_T^i	total electron impact ionization cross section
σ_T^{ei}	total electron-ion recombination cross section
σ_T^s	total scattering cross section
$\bar{\tau}_i$	average ion lifetime
τ_{min}	minimum greyscale intensity time
τ_p	pixel dwell time
τ_L	line scan time
υ	fraction of SEs escaping back diffusion
Φ	work function
$\Phi_{hv}(\lambda)$	radiant photon flux
χ	metastable coefficient
ψ	ionization rate
ω_{BSE}	BSE excitation efficiency
ω_{PE}	PE excitation efficiency
Ω	scattering solid angle
$\bar{\Omega}$	average SE-ion capture probability

Abstract

This thesis quantitatively investigates gaseous electron-ion recombination in an environmental scanning electron microscope (ESEM) at a transient level by utilizing the dark shadows/streaks seen in gaseous secondary electron detector (GSED) images immediately after a region of enhanced secondary electron (SE) emission is encountered by a scanning electron beam. The investigation firstly derives a theoretical model of gaseous electron-ion recombination that takes into consideration transients caused by the time constant of the GSED electronics and external circuitry used to generate images. Experimental data of pixel intensity versus time of the streaks is then simulated using the model enabling the relative magnitudes of (i) ionization and recombination rates, (ii) recombination coefficients, and (iii) electron drift velocities, as well as absolute values of the total time constant of the detection system, to be determined as a function of microscope operating parameters. Results reveal the exact dependence that the effects of SE-ion recombination on signal formation have on reduced electric field intensity and time in ESEM. Furthermore, the model implicitly demonstrates that signal loss as a consequence of field retardation due to ion space charges, although obviously present, is not the foremost phenomenon causing streaking in images, as previously thought.

Following that the generation and detection of gaseous scintillation and electroluminescence produced via electron-gas molecule excitation reactions in ESEM is investigated. Here a novel gaseous scintillation detection (GSD) system is developed

to efficiently detect photons produced. Images acquired using GSD are compared to those obtained using conventional GSED detection, and demonstrate that images rich in SE contrast can be achieved using such systems. A theoretical model is developed that describes the generation of photon signals by cascading SEs, high energy backscattered electrons (BSEs) and primary beam electrons (PEs). Photon amplification, or the total number of photons produced per sample emissive electron, is then investigated, and compared to conventional electronic amplification, over a wide range of microscope operating parameters, imaging gases and photon collection geometries. The main findings of the investigation revealed that detected electroluminescent signals exhibit larger SE signal-to-background levels than that of conventional electronic signals detected via GSED. Also, dragging the electron cascade towards the light pipe assemblage of GSD systems, or electrostatic focusing, dramatically increases photon collection efficiencies. The attainment of such an improvement being a direct consequence of increasing the 'effective' solid angle for photon collection.

Finally, in attempt to characterize the scintillating wavelengths arising from sample emissive SEs, PEs, BSEs, and their respective cascaded electrons, such that future photon filtering techniques can be employed to extract nominated GSD imaging signals, the emission spectra of commonly utilized electroluminescent gases in ESEM, such as argon (Ar) and nitrogen (N_2), were collected and investigated. Spectra of Ar and N_2 reveal several major emission lines that occur in the ultraviolet (UV) to near-infrared (NIR) regions of the electromagnetic spectrum. The major photon emissions discovered in Ar are attributed to occur via atomic de-excitation transitions of neutral Ar (Ar I), whist for N_2, major emissions are attributed to be a consequence of second positive band vibrational de-excitation reactions. Major wavelength intensity versus gas pressure data, for both Ar and N_2, illustrate that wavelength intensities increase with decreasing pressure. This phenomenon strongly suggesting that quenching effects and reductions in excitation mean free paths increase with imaging gas pressure.